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Performance evaluation of remediation scenarios for DNAPL contaminated groundwater using analytical models and probabilistic methods

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Abstract

Chlorinated solvents are widespread groundwater contaminants. Treatment or removal of source areas and treatment of the groundwater plume are the two primary strategies used to reduce risks at contaminated sites. Optimal design of groundwater remediation strategies is important to maximize contaminant removal and reduce costs. We used an analytical model and probabilistic methods to evaluate the performance of remedial methods for groundwater contaminated by chlorinated solvents, evaluating different combinations of source removal and plume treatment. The results indicated that natural attenuation alone cannot control dissolved 1,1,1-trichloroethane plumes within the boundaries of the site, with the duration of the plume exceeding 60 years. When 20–80% of the source area was removed, the dissolved plume migrated beyond the site and the plume could not be contained within 50 m down gradient over 20 years. Without source removal, the dissolved plume can only be restricted to within the site by enhanced remediation, and source removal reduces the duration of the dissolved plume. Treatment of the plume shortens the distance over which the dissolved plume travels. When source removal and treatment of the plume were jointly applied, the remediation efficiency of the contaminated groundwater was significantly improved. Analytical models and probabilistic methods have been proven to be very useful tools for optimizing the design of remediation strategies for NAPL-contaminated sites.

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1. Introduction

Chlorinated solvents are widespread groundwater contaminants often released as dense non-aqueous phase liquids (DNAPLs) (Stroo et al. 2012). In China, chlorinated solvents are also among the most frequently detected pollutants in groundwater at industrial contaminated sites. Chlorinated solvents are immiscible in water; however, their solubilities typically exceed regulatory values in groundwater. After being spilled into the environment, chlorinated solvents are frequently present as DNAPL, which serves as a concentrated source of groundwater contamination.

There are two primary zones at DNAPL sites, the source zone and the plume zone. Remediation strategies for DNAPL sites can be divided into two categories: source removal or treatment and contaminated plume cleanup. The main objective of source remediation is to reduce the mass, volume, or mobility of contaminants. Several technologies have been used to treat source areas, including in situ thermal treatment, in situ chemical oxidation (Seol et al. 2003), in situ chemical reduction (Soga et al. 2004), surfactant (or co-solvent) flushing (Mulligan et al. 2001), and in situ bioremediation. A performance evaluation of DNAPL source remediation using different technologies indicated that none of the treatment technologies was able to reduce concentrations below maximum contaminant levels throughout the source zone. The median concentration reduction of the chlorinated solvents was approximately 88% (McGuire et al. 2006).

It is more difficult to remediate residual contamination using any technology (Stroo et al. 2012). Complete removal of free-phase or residual NAPL is not possible due to the complex entrapment of NAPLs in the soil at field sites (Soga et al. 2004). The purpose of plume remediation is to reduce the area and contaminant concentrations of the dissolved plume. Approaches for remediating dissolved contaminant plumes including pump and treat (Rivett et al. 2006), enhanced bioremediation, permeable reactive barriers, and natural attenuation (McGuire et al. 2004). When only the plume is treated, the NAPL source remains in the ground and continues to release contaminants for a very long time.

Remedial technologies for source areas and dissolved contaminated plumes are generally expensive. It is often difficult at contaminated sites to determine whether remedial efforts should be focused on the contaminant source, the dissolved plume, or both (Falta 2008). To reduce the remediation cost, it is important to optimize the design of groundwater source and plume remediation strategies. Optimization of groundwater remediation activities involves maximizing contaminant removal while minimizing capital, operating, and maintenance costs (Finsterle 2006). Parker et al. (2010) investigated numerical optimization of DNAPL site remedial design considering the effects of prediction and measurement uncertainty. The results indicated that optimized designs yielded cost savings of up to ~50% compared with non-optimized designs based on common engineering practices. Cardiff et al. (2010) used a semi-analytical model to optimize DNAPL source and plume remediation strategies. Probabilistic methods, historical data, and prior information were combined to reduce cost optimization uncertainty. Falta et al. (2008) developed a new analytical model for simulating the transient effects of simultaneous groundwater source and plume remediation. This model allows for flexible simulation of natural attenuation and remediation efforts that enhance plume degradation.

The above studies indicated that mathematical models can play an important role in the remedy selection process. Many models can be used to optimize the design of remediation strategies for NAPL-contaminated sites; however, most of these models are highly complex and difficult to apply in practice. Therefore, analytical models such as BIOCHLOR, BIOSCREEN, etc., have been most frequently used for contaminated sites. In this study, an recently developed analytical model and probabilistic methods were used to simulate source removal and plume remediation at a chlorinated solvents-contaminated site in China. The main objective was to evaluate the performance of various scenarios for remediating contaminated groundwater.

2. Materials and Methods

2.1 Study area

The study area was a former mechanical plant located in eastern China with an area of 100,000 m², and it mainly engaged in the production of automotive air conditioning systems. Chlorinated hydrocarbons such as 1,1,1-trichloroethane (1,1,1-TCA) were used as cleaning agents from 1996 to 2006. About 240 t of 1,1,1-TCA were

used each year; therefore, a total of ~2400 t were consumed.

The geology underlying this site can be divided into three main units: (1) an upper zone of fill material with a thickness of 0.7–2.6 m; (2) a middle layer of clayey silt with a thickness of 0.8–4.0 m; and (3) a bottom layer of silty clay with a thickness of 8–9.5 m. This bottom confining layer limits the depth of contamination to the more permeable upper layers. The water table is 0.4–1.8 m below ground surface and the flow direction is generally from southeast to northwest. The horizontal hydraulic conductivity (K_H) ranges from 2.17×10^{-7} cm·sec⁻¹ (clay) to 2.56×10^{-4} cm·sec⁻¹ (clayey silt), with an average hydraulic conductivity of 2.0×10^{-6} cm·sec⁻¹. The vertical hydraulic conductivity (K_v) of the silty and clayey sands is 1.25×10^{-7} to 1.53×10^{-4} cm·sec⁻¹. Horizontal gradients vary between 0.001 and 0.004 m·m⁻¹. There are no significant seasonal variations in groundwater flow direction.

In 2004–2009, several site investigations were conducted. 1,1,1-TCA and its degradation products were found in the soil and groundwater. 1,1,1-TCA and 1,1-dichloroethane (1,1-DCA) were the most frequently detected contaminants in the soil, with maximum concentrations of 2410 and 384 mg·kg⁻¹, respectively. The most heavily contaminated region was located 6–10 m below the ground surface. In groundwater, the main contaminants were also 1,1,1-TCA and 1,1-DCA. The maximum and median concentrations of 1,1,1-TCA were 1,510,000 and 181 µg·L⁻¹, respectively. The maximum and median concentrations of 1,1-DCA were 382,000 and 147 µg·L⁻¹, respectively. The maximum concentrations of 1,1,1-TCA and 1,1-DCA exceeded the maximum acceptable level (intervention values suggested by Dutch standards) (Swartjes 1999).

To evaluate the ability of natural attenuation to reduce the contaminant concentrations in the groundwater plume, several groundwater monitoring wells were established and sampled. Hydrogeochemical indices (dissolved oxygen, redox potential, etc.), microorganisms that can degrade chlorinated hydrocarbons, and the biodegradation products of chlorinated hydrocarbons were also analyzed. The groundwater temperature was 15.4–28.8 °C. The pH was 5.89–12.06 with a mean pH of 7.58. Dissolved oxygen (DO) was 0.39–4.49 mg·L⁻¹ with a mean of 2.14 mg·L⁻¹. Several studies recorded DO concentrations <1 mg·L⁻¹, likely indicating anaerobic conditions in the groundwater (Clement et al. 2002). The oxidation-reduction potential (ORP) was -151 to -211 mV, with ORPs <300 mV indicating the presence of anaerobic conditions. The ORP of some samples was below 50 mV, indicating favorable conditions for reduction reactions. The anaerobic dechlorination bacteria *Dehalococcoides* spp. (qDHC) were detected in the whole samples. The final degradation products of 1,1,1-TCA (methane, ethylene, and ethane) were also detected in the groundwater samples. The above results indicate that natural attenuation of 1,1,1-TCA was occurring at the site.

2.2 Remediation evaluation model and remediation scenarios

2.2.1 Remediation evaluation model

The analytical model REMChlor was selected to evaluate remedial strategies for contaminated groundwater at the study area. The model was developed to simulate the transient effects of groundwater source and plume remedial efforts and focuses on defining the benefits of partial DNAPL source remediation (Falta 2008).

2.2.2 Remediation alternatives

Four remediation scenarios were established for this study, as follows:

Alternative 1: natural attenuation of the source zone and contaminated plume. The source zone was assumed to have no decay. The decay rates of 1,1-TCA and its biodegradation product in the plume (1,1-DCA) were assumed to be 1 yr⁻¹ and 0.2 yr⁻¹, respectively.

Alternative 2: source remediation and natural attenuation of the plume. Source remediation would be conducted for 5–7 years and 80% of the source mass would be removed. No remediation would be conducted for the contaminant plume. The decay rates of 1,1,1-TCA and 1,1-DCA were the same as for Alternative 1.

Alternative 3: contaminated plume remediation. Enhanced plume remediation would be conducted for 5–10 years. During this period, the decay rate of 1,1,1-TCA (10 yr⁻¹) and 1,1-DCA (2 yr⁻¹) would be 10 times higher than the natural attenuation rate. The enhanced remedial activity would only be applied to the plume 100 m downstream of the source.

Alternative 4: source remediation and enhanced plume degradation. Source remediation would be conducted for 5–7 years. The percent mass removal at the source was varied from 20–80%. Enhanced plume degradation would be conducted for 5–10 years. The decay rate of the TCA plume was varied between 1 and 10 yr⁻¹, while the

decay rate of DCA was varied between 1 and 2 yr⁻¹.

2.2.3 Probabilistic remediation evaluation

The Probabilistic Remediation Evaluation Model for Chlorinated Solvents (PREMChlor) was developed to simultaneously evaluate the effectiveness of source and plume remediation, including consideration of the uncertainties in all major parameters. PREMChlor was developed by linking the analytical model REMChlor to the Monte Carlo modeling package GoldSim via a FORTRAN dynamic-link library application (Liang et al. 2010). In PREMChlor, all of the uncertain input parameters are treated as stochastic parameters represented by probability density functions. The outputs from PREMChlor are probability distributions and summary statistics for the distributions. Cost analysis of common technologies for DNAPL source removal and dissolved plume treatment are included. PREMChlor gives users a single platform in which cost, source treatment, plume management, monitored natural attenuation, and risk assessment can all be evaluated at once, and in which uncertainty can be incorporated into the site decision-making process.

2.2.4 Parameters values for the model

Input parameters used in REMChlor were based on available site data. Where site-specific data were not available, reasonable assumptions were made based on widely accepted values. The site was modeled using a grid size of 75 rows by 30 columns. Each grid cell was 20 m long by 10 m wide. The main parameter values are listed in Table 1.

Table 1 Parameter value of the remediation evaluation model and probabilistic remediation evaluation				
Parameter	Unit	Value	Probability distribution	Data acquired
Source parameters				
Initial source zone contaminant mass	kg	20000	Deterministic value	Field investigation
initial source zone concentration	g·L ⁻¹	1.5	Deterministic value	Field investigation
power function exponent		1	Deterministic value	Empirical value
Source zone width	m	40	Deterministic value	Field investigation
Source zone depth	m	10	Deterministic value	Field investigation
Source removal ratio		0.2-0.8	Triangular Distribution	
Transport parameters				
Darcy velocity	m·yr ⁻¹	8	Deterministic value	Field investigation
Effective porosity		0.4	Deterministic value	Field investigation
Retardation factor		2	Deterministic value	Empirical value
Source decay rate	yr ⁻¹	0	Deterministic value	Empirical value
Coefficient of variation for velocity field		0.1	Deterministic value	Empirical value
Minimum normalized streamtube velocity		0.5	Deterministic value	Empirical value
Maximum normalized streamtube velocity		1.5	Deterministic value	Empirical value
Transverse dispersivity		0.2	Deterministic value	Empirical value
Vertical dispersivity		0.05	Deterministic value	Empirical value
Plume decay rates				
1,1,1-TCA	yr ⁻¹	1-10	Triangular Distribution	Literature value
1,1-DCA	yr ⁻¹	0.2-2	Triangular Distribution	Literature value

3. Results and discussion

3.1 Alternatives 1 and 2: source remediation with natural attenuation of the plume

Contaminant concentration contours for TCA under Alternative 1 (no source removal) and Alternative 2 (source removal of 80%) are shown in Fig. 1. The TCA plumes were delineated to the maximum contaminant level (300 µg·L⁻¹). After 10 years, the TCA-contaminated plume extended about 120 m downgradient under both scenarios (Fig. 1). The leading edges of both plumes extended beyond the boundaries of the site (100 m downgradient of the source). The plume was approximately 40 m wide under both scenarios. With no source

removal (Alternative 1; Fig. 1a), the TCA concentration declined in the downgradient direction and the highest concentration ($>1.2 \times 10^5 \mu\text{g}\cdot\text{L}^{-1}$) was observed near the source location. With 80% removal of the source (Alternative 2; Fig. 1b), there were two obvious high-concentration regions, one near the source and the other 40 m downgradient. The highest concentration ($4.3 \times 10^4 \mu\text{g}\cdot\text{L}^{-1}$) was significantly lower than the highest concentration with no source removal. Twenty years later, under Alternative 1, the leading edge of the groundwater contaminated plume had migrated 140 m downgradient and the maximum plume width had been reduced to only 31 m. The maximum contaminant concentration in the plume had also declined to $1.3 \times 10^4 \mu\text{g}\cdot\text{L}^{-1}$. Under Alternative 2, the contaminated plume had migrated 96 m downgradient and was 29 m wide. The maximum concentration was $4.3 \times 10^3 \mu\text{g}\cdot\text{L}^{-1}$. In general, compared with Alternative 1, the plume area under Alternative 2 was smaller and the concentrations within the plume lower. Importantly, the plume was also contained within the site boundaries.

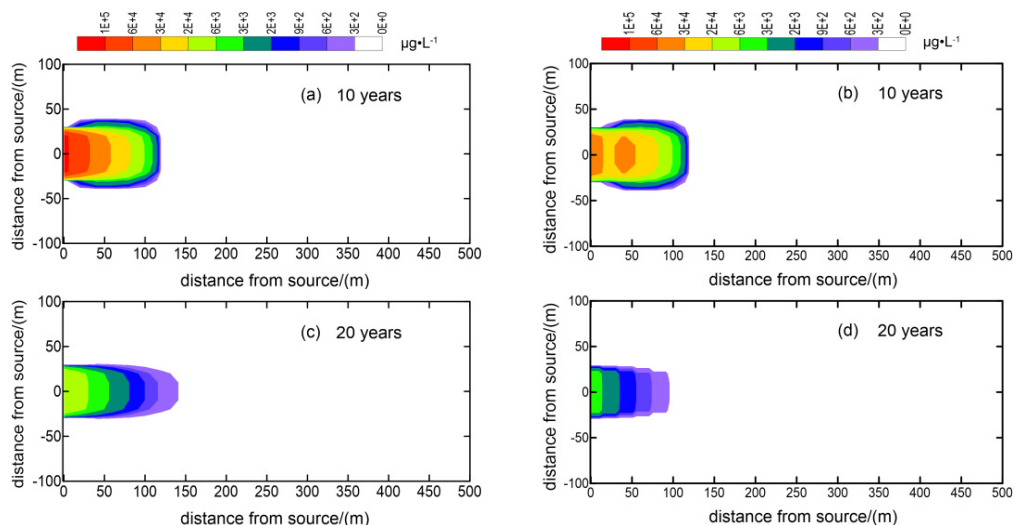


Fig. 1. Concentration contours for TCA plumes. (a) and (c) Alternative 1: no source removal, (b) and (d) Alternative 2: 80% source removal, (a) and (b) after 10 yr, and (c) and (d) after 20 yr.

Under anaerobic conditions, the main biodegradation product of 1,1,1-TCA is 1,1-DCA (Scheutz et al. 2011). The predicted plumes for 1,1-DCA are shown in Fig. 2. According to the simulation results, under Alternatives 1 and 2, the groundwater plumes remained above the maximum contaminant level for >40 years. After 10 years, the DCA plume was located 10–139 m downgradient. The leading edge of the plume extended beyond the site boundaries. After 20 years, the DCA plume had migrated to 256 m downgradient. After 40 years, the entire DCA plume had migrated outside of the site; the leading edge of plume was 457 m downgradient. The spatial distributions of DCA in the plumes were similar at different times; along the flow direction of the groundwater, the DCA concentrations first increased and then decreased, with the downgradient portions of the plumes having higher contaminant concentrations. After 10 years, the length of the DCA plume was approximately equal to that of the TCA plume. After 20 years, the length of the DCA plume was 1.8 times that of the TCA plume, indicating that DCA was the main contaminant in plume. At 10 and 20 years, the size of the DCA plumes under Alternatives 1 and 2 were similar. However, after 40 years, the size of the DCA plume under Alternative 2 (source removal) was clearly smaller than that under Alternative 1 (no source removal) and the contaminant concentrations were also significantly lower.

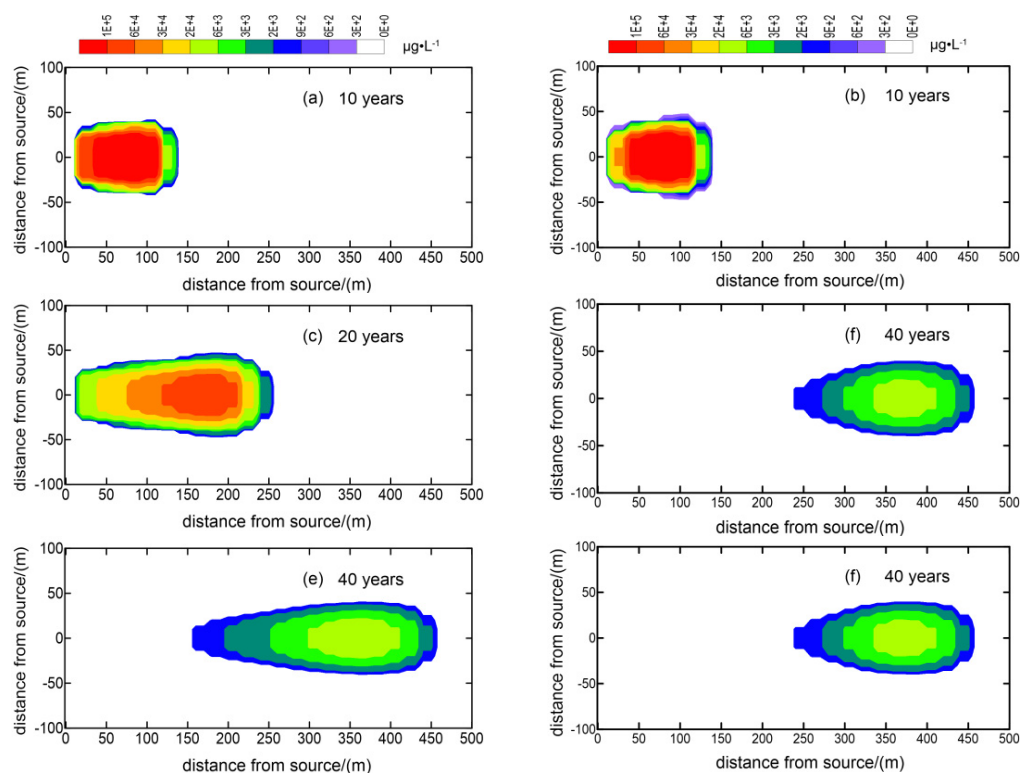


Fig. 2. Concentration contours for DCA plumes. (a), (c), and (e) Alternative 1: no source removal, (b), (d), and (f) Alternative 2: 80% source removal, (a) and (b) after 10 yr, (c) and (d) after 20 yr, (e) and (f) after 40 yr.

To evaluate the impact of various degrees of source removal on the plume, PREMChlor was applied to predict the probability distribution of the contaminant concentrations downgradient. The percent source removal was varied over 20–80%. The amount of NAPL source removal that can be achieved under field condition is influenced by several factors, including soil properties, hydrogeology, NAPL distribution, etc. (Suchomel and Pennell 2006). Therefore, it is usually impossible to completely remove the NAPL source from the site. Previous studies have indicated that up to 60–90% removal can be achieved (McGuire et al. 2006). In the present study, we set the maximum source removal to 80%. The maximum migration distance and the longevity of the dissolved plume were the main indicators used to evaluate the efficiency of plume control under different source removal scenarios. The maximum acceptable migration distance of the plume was set to 100 m downgradient (the site boundary). If contaminants exceeded that distance, the remediation option was considered ineffective. The longevity of the plume was assessed 50 m downgradient of the source, as an indicator of the overall longevity of the plume.

In the simulations, at 50 m downgradient, the contaminant (1,1,1-TCA and 1,1-DCA) was first detected after 4 years. The concentration increased rapidly and reached a maximum at 6 years, after which the concentration decreased gradually (Fig. 3a). Between 4–9 years, the contaminant concentrations of the plumes were nearly the same for different source removal percentages. After 9 years, the differences between the concentrations of contaminant in the plumes increased and peaked at 12 years. The difference between the 5% and 95% predicted concentration was $97185 \mu\text{g}\cdot\text{L}^{-1}$. After 20 years, the contaminant concentrations were all within the range $10222\text{--}28998 \mu\text{g}\cdot\text{L}^{-1}$. Therefore, based on these results, it seems implausible to constrain the plume to within 50 m downgradient within 20 years. At least 35–40 years would be needed for the plume to be contained within 50 m.

At 100 m downgradient, the contaminant was first detected after 7 years. The concentration increased rapidly and reached a maximum at 11 years, after which the concentration gradually decreased (Fig. 3b). Between 7–11 years, the contaminant concentrations of the plumes were nearly the same for different source removal percentages. After 11 years, the differences between the concentrations of contaminant in the plumes increased and reached a

peak at 18 years. The difference between the 5% and 95% predicted concentration was $48758 \mu\text{g}\cdot\text{L}^{-1}$. After 8 years, the contaminant concentration of the plume ($23461 \mu\text{g}\cdot\text{L}^{-1}$) was higher than the maximum acceptable level ($300 \mu\text{g}\cdot\text{L}^{-1}$), indicating that the plume had migrated beyond the site boundaries. After 20 years, the 5% predicted contaminant concentrations was $21892 \mu\text{g}\cdot\text{L}^{-1}$. Therefore, under these source removal scenarios, the duration of the plume was longer than 20 years.

The above results indicate that removal of the source alone does not result in a rapid decrease in the concentration of the contaminated plume. In our study, the biodegradation rate of the contaminant and the groundwater flow rate were assumed to be constant. Therefore, the concentration of the contaminant in the plume is directly related to the release characteristics of the source. In REMChlor, the source model serves as a time-dependent mass flux boundary condition to the analytical plume model. The contaminant source model is based on a power function relationship between the source mass and source discharge. The exponent determines the shape of the source discharge response to changing source mass. In our study, the exponent = 1, indicating that a 1:1 relationship between the source mass and source discharge was assumed in the model. Under the source removal scenarios, the percent mass removal was varied between 20–80%; therefore, the dissolved concentration in the source zone differed among the source removal scenarios. Thus, lower source removal scenarios would result in higher groundwater concentrations and vice versa.

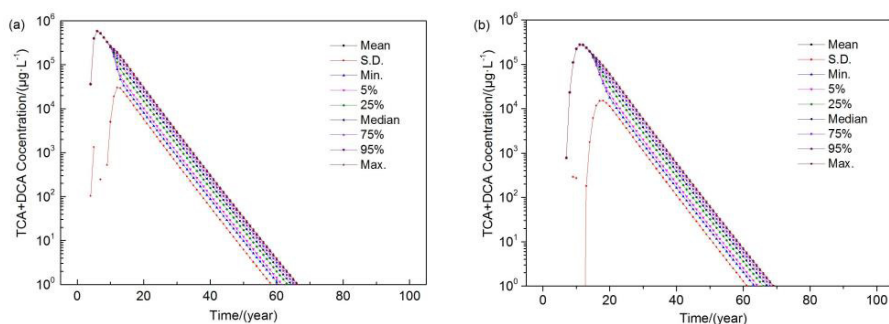
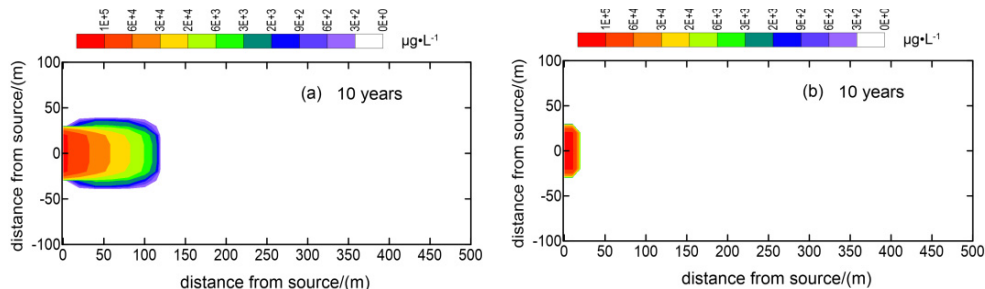


Fig. 3. The concentration of contaminants (1,1,1-TCA + 1,1-DCA) at (a) 50 m and (b) 100 m downgradient of the source with the percent source removal.

3.2 Alternative 3: enhanced plume remediation with no removal of the TCA source

To evaluate the impact of the decay rate on the plume behavior, two decay rates were selected. A low decay rate (1 yr^{-1}) was used to simulate natural attenuation (Alternative 1) and a high decay rate (10 yr^{-1}) was used to simulate enhanced plume remediation (Alternative 3). The predicted plumes are shown in Fig. 4. The TCA plumes were delineated to the maximum contaminant level ($300 \mu\text{g}\cdot\text{L}^{-1}$). Under enhanced plume remediation (Alternative 3), after 10 years, the plume extended 20 m downgradient, 1/6 as far as under natural attenuation. The highest concentration ($1.3 \times 10^5 \mu\text{g}\cdot\text{L}^{-1}$) was observed near the source location. After 20 years, the plume length was still 20 m, while that for natural attenuation was 140 m downgradient. The above results indicate that the contaminated plume can be contained within the site boundaries through enhanced plume remediation.



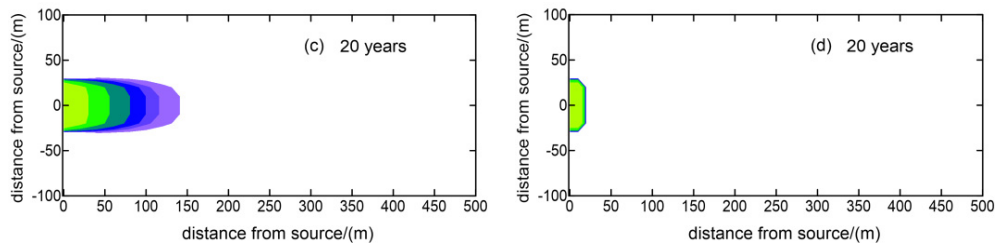


Fig. 4. Concentration contours of the TCA plumes after (a) and (b) 10 yr and (c) and (d) 20 yr with (a) and (c) decay rate = 1 yr^{-1} and (b) and (d) decay rate = 10 yr^{-1} .

The predicted DCA plumes are shown in Fig. 5. Under Alternative 3 (enhanced remediation), after 10 years, the plume extended 74 m downgradient, about 53% of the length of the plume under Alternative 1 (natural attenuation). Along the flow direction of the groundwater, the DCA concentration first increased and then decreased, with the highest concentrations in the center of the plume. The maximum concentration with enhanced plume remediation ($2.7 \times 10^4 \mu\text{g} \cdot \text{L}^{-1}$) was clearly lower than that under natural attenuation ($1.2 \times 10^5 \mu\text{g} \cdot \text{L}^{-1}$). After 20 years of enhanced remediation, the plume extended 38 m, about 15.6% of the length of the plume under natural attenuation. The concentrations in the plume under enhanced remediation were also significantly lower than those in the plume under natural attenuation. The predicted plumes of TCA and DCA under enhanced remediation had a maximum length of $<75 \text{ m}$; therefore, even without source removal, the plume could be contained within the site through enhanced plume remediation.

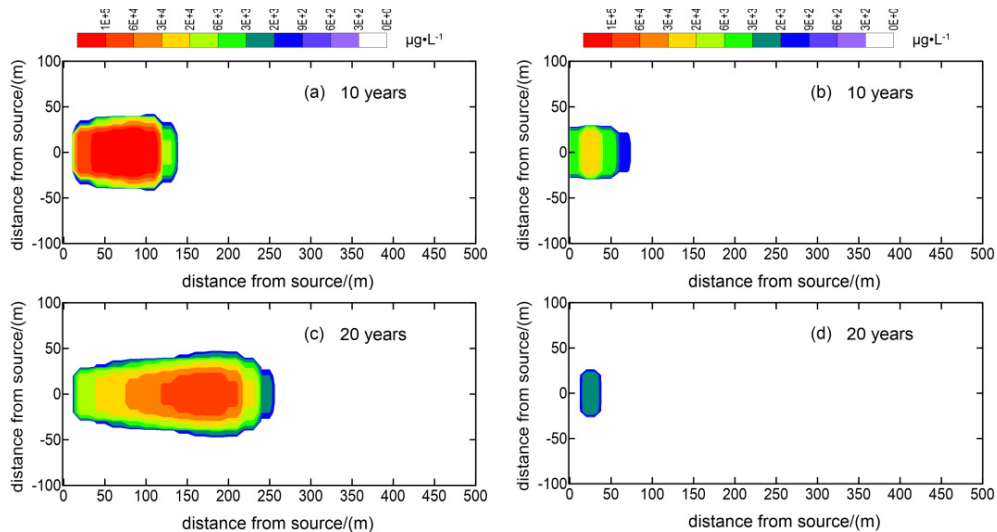


Fig. 5. Concentration contours of the DCA plumes after (a) and (b) 10 yr and (c) and (d) 20 yr with (a) and (c) decay rate = 1 yr^{-1} and (b) and (d) decay rate = 10 yr^{-1} .

To evaluate the impact of the decay rate on the behavior of the downgradient plume, the decay rates were assumed to vary over $1\text{--}10 \text{ yr}^{-1}$. At 50 m downgradient, the contaminant was detected after 4 years. The concentration increased rapidly and reached a maximum at 6 years, after which the concentration declined gradually (Fig. 6a). After 20 years, the contaminant concentrations were all within $771.3\text{--}22386 \mu\text{g} \cdot \text{L}^{-1}$ with varying plume decay rates. Therefore, it seems implausible to contain the plume within 50 m downgradient within 20 years; at least 25 years were estimated to be needed. The contaminated plume persisted for >48 years. At 100 m downgradient (the site boundary), the contaminant was detected at 7 years and reached a maximum at 10–12 years under various decay rates (Fig. 6b). The maximum concentrations of the plumes varied over $294.6\text{--}49788 \mu\text{g} \cdot \text{L}^{-1}$. Therefore, only when the decay rate is close to 10 yr^{-1} would the contaminated plume be contained within the site.

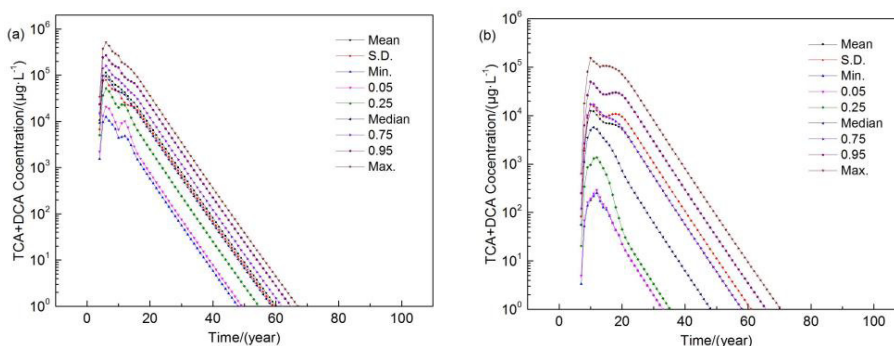


Fig. 6. Concentrations of contaminants (1,1,1-TCA + 1,1-DCA) (a) 50 m and (b) 100 m downgradient of the source zone with various plume decay rates.

3.3 Alternative 4: source removal with enhanced remediation of the dissolved plume

Joint application of source removal and enhanced plume remediation can improve the efficiency of contaminated groundwater remediation (Stroo et al. 2012). The performance of different combinations of source removal and enhanced plume remediation were evaluated. Source removal was varied over 20–80% and contaminant decay rates were varied over 1–10 yr⁻¹. At 50 m downstream, the contaminant was first detected at 4 years. The concentration increased rapidly and reached a maximum at 6 years, after which the concentration decreased gradually (Fig. 7a). After 20 years, contaminant concentrations were within 275.1–9145.4 µg·L⁻¹ for the different remediation scenarios. Therefore, it was possible to contain the plume to within 50 m downgradient within 20 years. At 100 m downgradient (the site boundary), the contaminant was first detected at 7 years and reached a maximum at 10–11 years under the various scenarios (Fig. 7b). The maximum concentrations of the plumes were within 180–36587 µg·L⁻¹. The probability of the maximum concentration in the dissolved plume being <300 is about 5%. At least 15–35 years was needed for the concentration in the dissolved plume to decrease below the regulatory value. The contaminated plume was predicted to persist for 27–61 years.

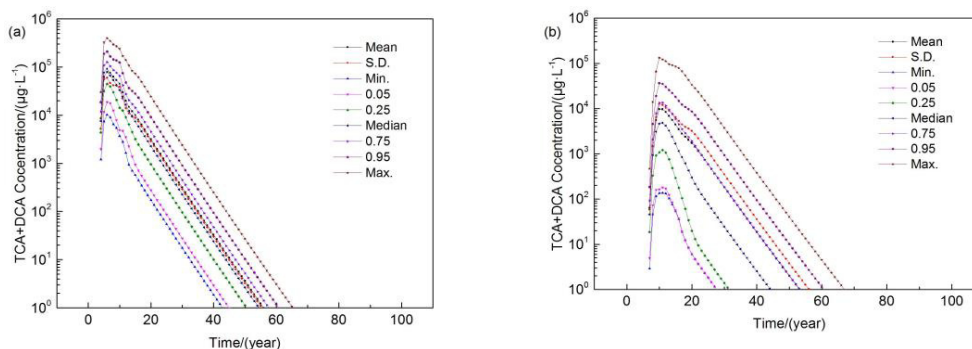


Fig. 7. Concentrations of contaminants (1,1,1-TCA + 1,1-DCA) at (a) 50 m and (b) 100 m downgradient of the source zone with various combination source removal and plume remediation scenarios.

4 Conclusions

The study area was heavily polluted by 1,1,1-TCA and its degradation products. The geochemical and hydrological conditions of the site were suitable for biodegradation of the pollutants by indigenous microbes present at the site. However, natural attenuation was not predicted to contain the dissolved TCA plume within the

boundaries of the site (100 m downgradient of the source); after 10 years, the plume was expected to migrate beyond the site. During the first 10 years, the main contaminant in the dissolved plume was 1,1,1-TCA. At 10 years, the size of the dissolved DCA plume was 1.8 times that of the TCA plume; therefore, DCA was expected to be the main contaminant in plume thereafter. When source removal was varied from 20% to 80%, the leading edge of the dissolved plume migrated beyond the site boundaries and the plume could not be contained within 50 m downgradient within 20 years. Without source removal, the dissolved plume could be contained within the site by enhanced remediation (decay rate close to 10 yr^{-1}). Source removal reduced the longevity of the dissolved plume and plume remediation shortened the migration of the dissolved plume downgradient. When source removal and plume remediation were jointly applied, the remediation efficiency of the contaminated groundwater significantly improved.

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References

- Cardiff M, Liu X, Kitanidis PK, et al. Cost optimization of DNAPL source and plume remediation under uncertainty using a semi-analytic model. *J Contam Hydrol* 2010; **113**:25-43.
- Clement TP, Truex MJ, Lee P. A case study for demonstrating the application of U.S. EPA's monitored natural attenuation screening protocol at a hazardous waste site. *J contam hydrol* 2002; **59**:133-162.
- Falta RW. Methodology for Comparing Source and Plume Remediation Alternatives. *Ground Water* 2008; **46**:272-285.
- Finsterle S. Demonstration of optimization techniques for groundwater plume remediation using iTOUGH2 *Environ Modell and Softw* 2006; **21**:665-680.
- Liang H, Falta RW, Newell CJ, et al. Decision & Management Tools for DNAPL Sites: Optimization of Chlorinated Solvent Source and Plume Remediation Considering Uncertainty. Clemson University. 2010.
- McGuire TM, McDade JM, Newell CJ. Performance of DNAPL Source Depletion Technologies at 59 Chlorinated Solvent-Impacted Sites. *Ground Water Monit Remed* 2006; **26**:73-84.
- McGuire TM, Newell CJ, Looney BB, Vangelas KM, Sink CH. Historical analysis of monitored natural attenuation: A survey of 191 chlorinated solvent sites and 45 solvent plumes. *Remediation J* 2004; **15**:99-112.
- Mulligan CN, Yong RN, Gibbs BF. Surfactant-enhanced remediation of contaminated soil: a review. *Eng Geol* 2001; **60**:371-380.
- Parker J, Kim U, Kitanidis P K, et al. 2010. Stochastic Cost Optimization of Multistrategy DNAPL Site Remediation. *Ground Water Monit R* 2010; **30**: 65-78.
- Rivett MO, Chapman SW, Allen-King RM, et al. Pump-and-Treat Remediation of Chlorinated Solvent Contamination at a Controlled Field-Experiment Site. *Environ Sci Technol* 2006; **40**:6770-6781.
- Scheutz C, Durant ND, Hansen MH, et al. Natural and enhanced anaerobic degradation of 1,1,1-trichloroethane and its degradation products in the subsurface – A critical review. *Water Res* 2011; **45**:2701-2723.
- Seol Y, Zhang H, Schwartz FW. A Review of In Situ Chemical Oxidation and Heterogeneity. *Environ Eng Geosci* 2003; **9**:37-49.
- Soga K, Page JWE, Illangasekare TH. A review of NAPL source zone remediation efficiency and the mass flux approach. *J hazard mater* 2004; **110**:13-27.
- Stroo HF, Leeson A, Marqusee JA, et al. Chlorinated Ethene Source Remediation: Lessons Learned. *Environ Sci Technol* 2012; **46**:6438-6447.
- Suchomel EJ, Pennell KD. Reductions in Contaminant Mass Discharge Following Partial Mass Removal from DNAPL Source Zones. *Environ Sci Technol* 2006; **40**:6110-6116.
- Swartjes F. Risk-Based Assessment of Soil and Groundwater Quality in the Netherlands: Standards and Remediation Urgency. *Risk Anal* 1999; **19**:1235-1249.